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Josephson effect through a ferromagnetic layer

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Abstract. We report transport measurements on Superconductor/Ferromagnet/Superconductor (S/F/S) junctions: Nb/Al/Gd/Al/Nb where gadolinium (Gd) is a weakly polarized ferromagnet. A sizeable critical current I_c is observed in the $I(V)$ characteristics. This current can be modulated by a weak magnetic field, as expected for a Josephson current. With these experiments, we establish that superconducting coherent transport survives across a small ferromagnetic layer. The penetration depth of Cooper pairs in Gd has been measured. An extensive study of the Josephson critical current in temperature for different thicknesses of magnetic compounds is presented. A comparison of transport measurements with S/N/S junction is given through measurements made on Nb/Al/Y/Al/Nb, where yttrium (Y) is used as non magnetic rare earth metal.

PACS. 74.50.+r Proximity effects, weak links, tunneling phenomena, and Josephson effects – 74.80.Dm Superconducting layer structures: superlattices, heterojunctions, and multilayers – 75.70.Cn Interfacial magnetic properties (multilayers)

1 Introduction

Interplay between superconductivity and magnetism has been a long standing debate in solid state physics. Indeed, since the ground state of a BCS superconductor consists of Cooper pairs with electrons in a singlet configuration, we do expect the strong exchange field of a ferromagnet to weaken the superconducting state. Competition and coexistence between these two orders have been studied in ternary compounds [1], but the way a Cooper pair passes through a bulk ferromagnetic layer remains poorly understood. Many experiments were done on S/F superlattices providing information on transition temperature in such multilayers and hence on penetration depth of Cooper pairs in the F layer indirectly. An other way to improve our knowledge of interplay between superconductivity and magnetism consists of studying the proximity effect between a superconductor and a metallic ferromagnet in S/F/S junction, and providing evidence of Josephson coupling through a ferromagnetic layer.

When a superconducting reservoir S is connected through good electrical contacts to a normal metal N, Cooper pairs propagate in the latter keeping their coherence over a characteristic length ξ_N, which is usually fixed by the temperature. In case of a ferromagnet F, the exchange field J strongly dephases the Cooper pairs [2]. This has two consequences. First, the coherence length ξ_F is thus fixed by J, and expected to be very small [3,4]. Second, the superconducting order parameter is an oscillating function of the penetration depth in F, since its phase is modulated by J [5,6]. Therefore, the coupling between two superconducting layers through a ferromagnetic one may lead to new phenomena, as for example a non-monotonic behavior of the transition temperature *versus* the thickness of F (d_F) in S/F superlattices [6,7]. Experimental works supporting this scenario have been published recently [8,9], but those results are still controversial [10,11] because many different parameters can account for such non-monotonic decreases as for instance growth technique, roughness of the layer, or change in the magnetic behavior with the thickness of F.

Instead of using macroscopic measurements like the resistive transition temperature of multilayers, we designed a series of experiments to directly study the proximity effect in S/F/S junctions. The quantum coherence is probed by Josephson measurements. The critical current is expected to decrease non-monotonically with d_F and to change sign for given values of d_F. This leads to the so-called π-junction first predicted by Bulaevskii *et al.* in 1977, since a π phase shift is added in the standard Josephson relation [12,6]. Such π-junctions have been observed in High T_c superconductors [13,14], due to intrinsic

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phase shifts in the d-wave order parameter. Furthermore, negative critical currents have been observed in out of equilibrium low T_c compound [15], but never negative critical currents were evidenced in S/F systems. In this article we report on Josephson coupling between two BCS superconductors. An extensive study of the temperature and magnetic field dependence of the critical current, as well as the dependence *versus* the thickness of the ferromagnetic compound is presented.

2 Experimental details

2.1 Experimental set-up

All previous experiments on S/F systems where made by transport measurements on superlattices with the current running in the plane (CIP technique). We used a more relevant geometry to probe the coupling of S through F; the current perpendicular to the plane technique (CPP) [16]. Using this method we can be sure that the current passes through all the layers. This is not the case in the CIP method where it is very difficult to control specular reflection at the interface between the F and S layer. Since ξ_F is expected to be a few nanometers, the typical d_F has to be of the same order of magnitude. Thus minute resistances in the normal state of S/F/S junctions are expected requiring a specific high sensitive technique for the measurements.

A dc-current method based on superconducting choppers has been used to measure voltage down to picovolt [16] scale in a dilution refrigerator down to 50 mK. Therefore resistances in the nano-Ohm range can be measured. The actual S/F/S junctions are: Nb/Al/Gd/Al/Nb, using gadolinium as the ferromagnet. Supercurrent measurements will be compared to Josephson measurements on S/N/S junctions like: Nb/Al/Y/Al/Nb, where Y is used as a non magnetic rare earth metal. The superconducting layers are in fact Nb-25 nm/Al-150 nm sandwich. Its T_c being a little bit lower (around 8 K) than the one of bulk Nb (around 9 K), the presence of the aluminium layer avoids spurious effects coming from the superconducting Nb leads transition. Indeed, the presence of a second superconductor with a lower T_c allows us, as seen in Figure 1a, to separate the transition of the leads (Nb) around 8 K and the transition of the actual junction Al/Gd/Al, which depends on the bias current. The Gd thickness ranges from 2 to 10 nm in our sample. A schematic cross section of the sample is given in the inset of Figure 1.

2.2 Sample preparation

The layers are prepared by e-beam evaporation in ultralhigh vacuum on (100) silicon substrate held at room temperature using five different mechanical masks to pattern the junction. Evaporated thicknesses are measured with a quartz crystal microbalance. A typical rate of 0.1 nm/s is used for Gd. The accuracy of the Gd thickness measured by quartz crystal, estimated to be 0.25 nm, has been checked by Rutherford Backscattering Spectrometry (RBS) *a posteriori*. The base pressure in the deposition chamber is 10^{-9} torr and the working pressure is 2×10^{-9} torr for Gd and 4×10^{-9} torr for Nb and Al. The junction area (10^{-2} mm^2) is defined with an evaporation of an insulator (50 nm of SiO) through a shadow mask at 2×10^{-8} torr.

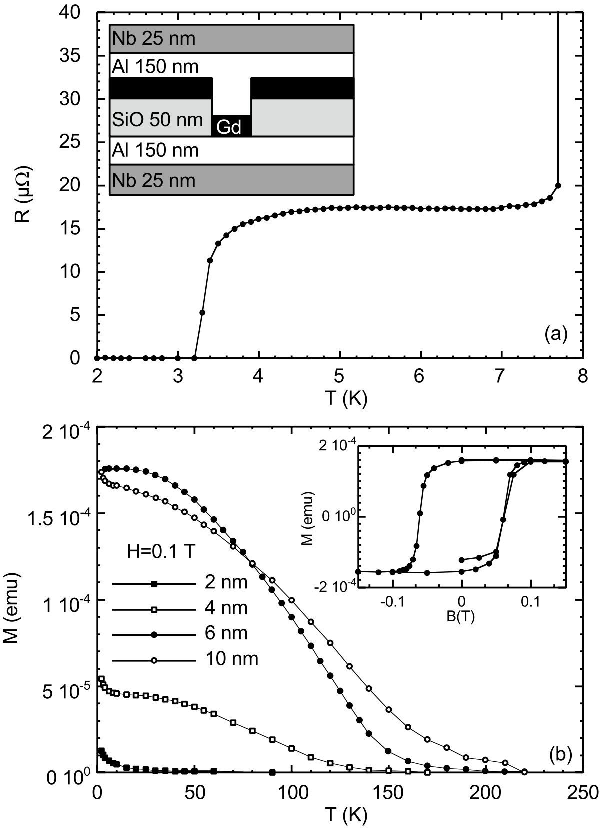

Fig. 1. (a) Superconducting characterization of the Nb/Al layer by resistance *versus* temperature measurements for a junction with a layer of Gd of 4 nm using a bias current of 1 mA. A schematic sample cross section is shown on the inset. (b) Magnetization *versus* temperature for different layer of gadolinium at $H = 0.1$ T. The inset shows an hysteresis loop measured on a layer of 6 nm of Gd.

2.3 Sample characterization

The first stage was the characterization of the superconducting behavior of the samples. Figure 1a displays the resistance of the junction containing 4 nm of Gd as a function of the temperature for a bias current (I_B) of 1 mA. Three different regimes can be distinguished separated by two different transition temperatures. In the first regime between 0 and 3.2 K, the junction is completely superconducting by proximity effect. A current can go through the ferromagnetic layer without voltage across the junction. The first transition temperature at 3.2 K defines the second regime, where the junction is resistive. Finally the second transition related to the transition of the superconducting Nb leads defined the last regime where all the compounds are in the normal state. The 7.8 K transition does not change with the bias current, but the first transition is strongly I_B dependent which confirms that it is related to the Gd junction itself.

Since we are dealing with low thickness ferromagnetic layers, it is of great importance to characterize the actual structure and magnetic behavior of our Gd films. Transmission Electron Microscopy (TEM) plane views of Al/Gd reference sample evaporated under the same conditions than the junctions showed that Gd layers thicker than 2 nm should be likely continuous as expected [17], giving a lower bound of Gd thicknesses used in this work. By working with junctions with more than 2 nm of Gd, we can expect that the ferromagnetic layers in the junction are continuous. From the TEM measurements, Gd layers appear to be polycrystalline, with a typical grain size of 7 nm.

The magnetic properties of Gd were investigated by means of SQUID magnetometry. Figure 1b displays the magnetization as the function of the temperature $M(T)$ for different Gd thicknesses with a field of 100 mT. The magnetic field is applied in the plane of the film. These curves show that the layers are indeed ferromagnetic, but not with bulk Gd characteristics, *i.e.* a Curie temperature of 289 K and a magnetic moment per atom of $7.6\mu_B$. The estimated Curie temperatures range from 50 to 150 K, and increase with the Gd thickness. From the saturation magnetization at low temperature (see the square hysteresis loop in the inset Fig. 1b), the magnetic moment per atom is estimated to be roughly $3.5\mu_B$, almost independent of the Gd thickness (see inset Fig. 6). This behavior can be understood if we consider a granular ferromagnet (*cf.* TEM measurements). The reduction of the moment can be attributed to frozen spin that do not align in the applied magnetic field due to spin glass effect as it is expected for very small Gd grains [18]. This reduction can also be the consequence of the existence of an angle between the easy axis and the applied magnetic field [19]. The $M(T)$ curve shape and the low Curie temperature correspond to the super-paramagnetic behavior known in low thickness Gd layers [20]. The inset of Figure 1b shows a typical magnetization curve $M(H)$ of Gd films. The coercitive field is around 100 mT independent of the layer thickness, and a significant remnant magnetization can be seen indicating the strong ferromagnetic behavior.

3 Results

We now have a good understanding of the structure and magnetic behavior of the ferromagnetic Gd layer. Results of transport measurements made with the dc-current method already described can be presented. A typical current-voltage characteristic is shown in Figure 2. For Gd thickness between 2 and 10 nm, a well defined critical current is always observed in our S/F/S junctions at low temperature. The linear shape of the I-V above the critical current is indicating an ohmic behavior. Therefore, we can use the Resistively Shunted Junction (RSJ) model assuming that the current is a Josephson current to fit the $I(V)$ curve, through the equation:

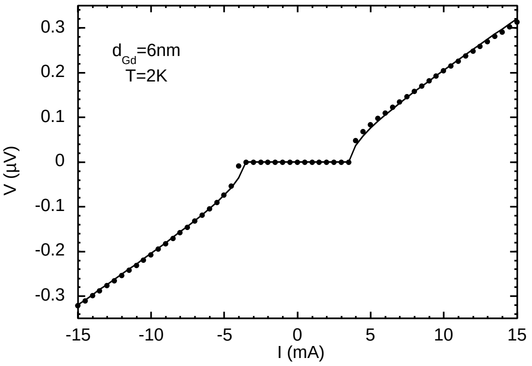

Fig. 2. Current-voltage characteristic of a junction containing 6 nm of Gd at 2 K. The critical current of 3.65 mA is well defined and an ohmic behavior is observed ($R = 2.2 \times 10^{-5}\ \Omega$). The solid line represents the RSJ fit (see the text), which is in good agreement with the data.

$$V = R\sqrt{I^2 - I_c^2}$$

where V is the voltage, R the normal state resistance and I_c the critical current. As seen by the agreement between the solid line and the data in Figure 2, such a Josephson based relationship accounts for the experiments. This is the first argument in favor of a Josephson coupling through a ferromagnetic Gd layer in our junctions.

In such UHV evaporated junctions, resistances remain rather high (around $10^{-5}\ \Omega$) instead of resistances of the order of $10^{-7}\ \Omega$ expected for such junctions with Gd assuming the resistivity for Gd at 140 $\mu\Omega$ cm. The presence of low transparency in the junction could arise due to adding factors such as bad interface quality (grain boundaries, partial oxidation), Fermi wave vector mismatch between Al and Gd [21] or due to the ferromagnetic character of Gd (spin dependent potential barrier etc.). The resistive behavior of such S/F/S junctions was extensively studied in an anterior work [22,23]. Because the potential barrier between the S and F layers plays a crucial role in the proximity effect [24,25], such a low interface transparency will be of great importance for the interpretation of the results. Resistance values of such junctions are spread over few orders of magnitude. But for the study of critical current *versus* Gd thickness, only junctions in the same range of resistance will be considered (around $5 \times 10^{-5}\ \Omega$).

The temperature variations of the critical current are in perfect agreement with classical metallic junctions. The Figure 3 shows the temperature dependence for two different samples with 3 nm and 4 nm thickness of Gd in the junction.

Both samples show an increase of critical current by decreasing the temperature with a positive curvature, which is the signature of metallic contact in the junctions. The sample with 4 nm of Gd exhibits a saturation of the increasing critical current at low temperature. In the case of the 3 nm of Gd sample, the critical current was too high to be measured at low temperature. But for samples

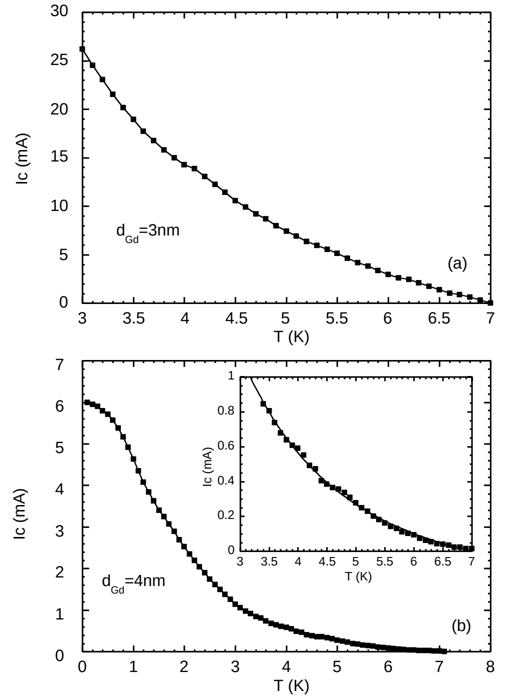

Fig. 3. Critical current *versus* temperature for two samples: (a) in the case of a junction containing 3 nm of Gd, the behavior close to T_c is linear; (b) the second junction with 4 nm of Gd shows a strong increase of critical current at low temperature with a positive curvature. The behavior close to T_c is shown in the inset.

with thicker layer of Gd, such a saturation was observed. Usually, the behavior close to T_c is rich in information. For an S/N/S junction we expected a quadratic behavior for $I_c(T)$ [26], meanwhile a linear shape is expected in the case of S/F/S junction [27]. In Figure 3a the linear behavior is much clearer than in the case of 4 nm of Gd junction (see the inset of Fig. 3b).

Most samples showed a rather clear linear behavior close to T_c. Unfortunately, accuracy on critical current *versus* temperature measurements near T_c is not sufficiently satisfactory to make a indisputable discrimination between a linear (S/F/S) and a quadratic (S/N/S) behavior of $I_c(T)$. Thus the signature of a ferromagnetic layer is weak in $I_c(T)$ measurement.

From the study of *I-V* characteristics and $I_c(T)$, there are several clues for the existence of a Josephson current due to superconducting proximity effect in the ferromagnetic layer. Furthermore, investigation of critical currents with weak applied magnetic fields will be justified.

Critical current measurements under a weak magnetic field H applied in the junction plane were performed for each thickness of Gd in the junctions. As seen in Figure 4, modulation of I_c can be obtained as a function of H, as expected from the Josephson relationship. Although the Fraunhofer patterns are strongly perturbed (see Fig. 4a) a periodicity can be extract between 0.2 to 0.6 mT depending on the sample corresponding approximately to the penetration of one magnetic flux quantum in the junctions. Figure 4b shows a SQUID like modulation of critical current attributed to strong edge currents in the sample with a thick layer of Gd in it (above 5 nm). Such measurements cannot completely exclude the presence of an array of tiny pinholes, but as we saw before, expecting continuity for Gd layer above 2 nm, such modulations are significantly relevant to prove the existence of a supercurrent through a ferromagnetic layer. In Figure 4a, the shape of the $I_c(H)$ deviates from the standard Fraunhofer pattern $sinc(x)$ expected for an homogeneous junction in an homogeneous field. In our case, for samples with a thin layer of Gd, the size of the junction is greater than the Josephson penetration depth λ_J, namely $\lambda_J = \sqrt{\phi_0/2\pi\mu_0 l J_c}$, where J_c is the critical current density, $l = 2\lambda_L + d_F$, λ_L is the London penetration depth (in Al: $\lambda_L = 50$ nm) and d_F the thickness of Gd, penetration of vortex in the junction is expected. In the case of $d_F = 2$ nm, we estimate λ_J to be 30 μm, smaller than $L = 100$ μm the size of the junction. Experiments at high temperature do not improve the quality of the $I_c(H)$ characteristics.

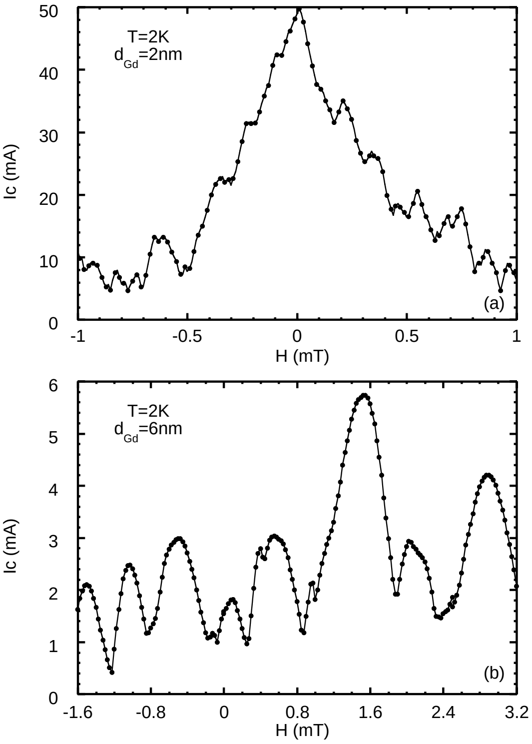

Fig. 4. Modulation of critical current *versus* an applied magnetic field in the plane of the junction for two samples: (a) in the case of a junction containing 2 nm of Gd, critical current can be modulated by an applied magnetic field; (b) this second junction with 6 nm of Gd shows also modulation with a rather visible periodicity of 0.5 mT.

Besides the fact that we deal with junctions in the large limit, the presence of a magnetic layer itself may strongly affect the $I_c(H)$ curve, leading to substantial perturbations in the distribution of critical currents, and hence to significant modifications of the usual Fraunhofer pattern. Those $I_c(H)$ curves were acquired before applying a strong magnetic field. It is of interest to compare measurements on magnetic rare-earth junctions with identical non-magnetic rare-earth junctions with yttrium, where

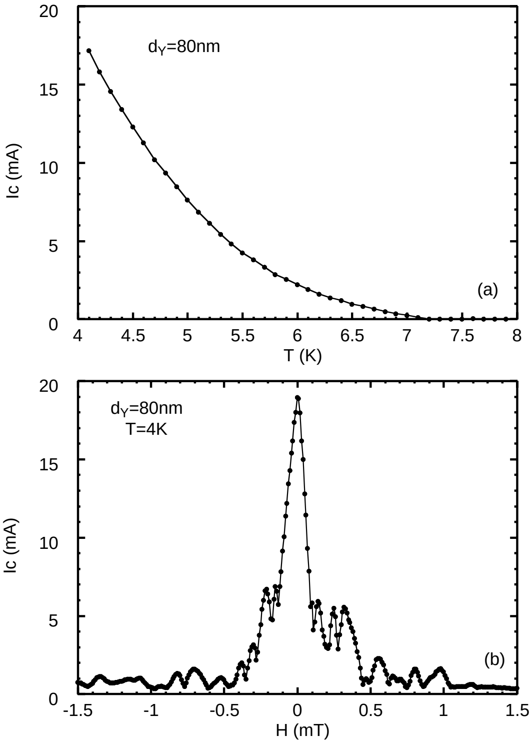

Fig. 5. Transport measurements on Nb/Al/Y/Al/Nb with $d_{\mathrm{Y}} = 80$ nm (a) $I_{\mathrm{c}}(T)$, the curve is concave as expected for an S/N/S junction with a quadratic behavior near T_{c}, (b) $I_{\mathrm{c}}(H)$ at 4 K. The modulation under magnetic field can be seen with a periodicity of 0.1 mT.

the geometry and superconductor layer thicknesses remain unchanged. Figure 5 shows results for $I_{\mathrm{c}}(T)$ and $I_{\mathrm{c}}(H)$ measurements obtained on a junction Nb/Al/Y/Al/Nb, with $d_{\mathrm{Y}} = 80$ nm. Because the proximity effect is much stronger in non magnetic metal, thicker film of yttrium were used compared to that with Gd to reduce the critical current. The variations of critical current near T_{c} is clearly quadratic, signature of S/N/S junction. As expected in case of non-magnetic metallic junction, the Fraunhofer pattern is much less perturbated. It is a clear indication that a large part of the deviations observed come from the magnetic behavior of the Gd layer.

Despite strong perturbations from the magnetic layer in the Fraunhofer pattern, we now have good evidence of Josephson effect through a ferromagnetic layer. Thus we can move to the central results of this paper. The aim of this work was to measure the penetration depth of Cooper pairs within the Gd layer (ξ_{F}). Figure 6 shows the Josephson critical current I_{c} as a function of the Gd thickness from 2 to 10 nm. In order to get an estimate of ξ_{F}, one needs to fit this curve with an appropriate theory. The occurrence of a supercurrent in S/N/S junctions relies on the quantum phase coherence between both superconducting sides of the junction. In normal metal, thermal energy controls the decoherence, and for diffusive metal the coherence length ξ_{N} is given by:

$$\xi_{\mathrm{N}} = \sqrt{\hbar D / 2\pi k_{\mathrm{B}} T} \tag{1}$$

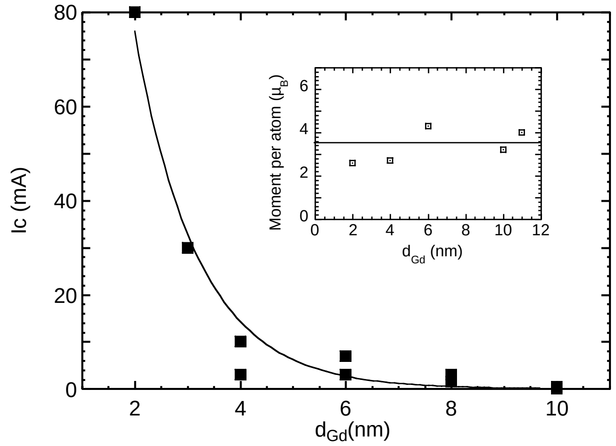

Fig. 6. Variation of the critical current *versus* Gd thickness at 1 K. The exponential law leads to a penetration depth of Cooper pairs of 1.2 nm. The magnetic moment per atom of Gd is independent of the thickness (insert).

with $D = v_{\mathrm{F}} l / 3$ the diffusion constant and T the temperature. I_{c} decreases exponentially near T_{c} with the normal thickness as $\exp(-d_{\mathrm{N}}/\xi_{\mathrm{N}})$ [28]. However, in the presence of an exchange field, the two electrons of opposite spins in Cooper pairs will have a different Zeeman energy. To keep the total energy constant, the spin up and down electrons will modify their kinetic energy, and therefore their speed.

As a result, Cooper pairs acquire a net moment and a severe dephasing process occurs as they evolve in a ferromagnetic layer [2]. In this case, the characteristic length ξ_{N} in the formula (1) is no longer relevant. The magnetic coherence length is given by [6,2]:

$$\xi_{\mathrm{F}} = \sqrt{4\hbar D / I} \tag{2}$$

with D the diffusion constant and I the exchange energy. For most ferromagnet, I is much greater than $k_{\mathrm{B}}T$ at low temperature so ξ_{F} is expected to be much smaller than ξ_{N}. Then the dominant depairing due to decoherence comes from the magnetic behavior of the Gd layer and not from temperature. In Figure 6, the line represents the fit to the data using the de Gennes-Werthamer theory in case of S/N/S junction. We can extract from this fit an experimental value of $\xi_{\mathrm{F}} = 1.2 \pm 0.2$ nm. Using formula (2), we can estimate the theoretical value for the magnetic coherence length $\xi_{\mathrm{F}} = 1.4$ nm, which is closed to the measured value, if we take an exchange energy of 250 meV for Gd, a Fermi velocity of 1.17×10^{7} cm/s and a mean free path on the order of the Gd thickness in the junction, *i.e.* $l = 5$ nm.

4 Conclusions

In this article, we showed that Josephson effect can be obtained through a strong ferromagnet. Despite the fact that Fraunhofer patterns are strongly perturbed, modulations of critical currents were obtained for low magnetic field applied on the junction. The comparison of the results with measurements on non magnetic rare-earth based junctions clearly indicates that perturbations come from the magnetic behavior of Gd. The variations of critical current with the thickness of the ferromagnetic layer were studied carefully. To prove the existence of a negative critical current in such junctions, a new experiment based on SQUID geometry is underway where one of the junction would contain magnetic impurities or magnetic compound, while the other one does not. If, as we expect, one of the critical current is negative, a phase shift could be observed in the $I_c(H)$ measurement.

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